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Construction of 6D Icosahedral Crystals from Approximate Icosahedral 3D Crystalline Structures and Phase Transitions

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Abstract.

From any so-called approximant 3D crystal, i.e. exhibiting approximate icosahedral units in real space and in reciprocal space, we build, using an optimal rational cut in $\mathbb{R}^6$, a 6D icosahedral crystal and the 3D i-phase associated with it. This construction becomes complete after "saturation" of the atom orbits under the action of a 6D icosahedral space group. Within the projection framework from $\mathbb{R}^6$, this predicts a splitting of the Wyckoff positions, in the 6D crystal, above each 3D Wyckoff position of the approximant crystal. Arithmetic selection rules, different from a band algorithm, are required to recover 3D approximant crystals by projection. No tiling of Euclidean physical space is considered here; only atom positions serve in the construction. Generalized order parameters can be defined in this scheme.

The icosahedral phase “i” coexists in several systems such as AlMnSi, TiNi, AlLiCu, with its “cousin” approximant crystalline phases of similar composition. In this study, we assume that the icosahedral quasicrystal and its cousin phases, in a given system, come from the same 6-dimensional icosahedral crystal, either by irrational projection in the first case or by double projection with a rational projector in the second case (Verger-Gaugry, 1990), within the projection framework (Duneau and Katz, 1985) from $\mathbb{R}^6$. We do not want to work with space tilings of 3D or 6D Euclidean space, but *directly* on the atomic positions of the approximant crystal, and we shall describe below how we are dealing with this choice. Since such 6D icosahedral crystals, when projected “irrationally” on physical space with a band algorithm (Gratias, 1988), provide icosahedral quasicrystals, we limit ourselves to the description of the method of construction of these 6D crystals. We shall describe in the following the construction method. At first, we set some requirements imposed by the projection method from $\mathbb{R}^6$ and indicate how optimal rational cuts in $\mathbb{R}^6$ are chosen with respect to local order in the approximant crystal.

ALGEBRAIC REQUIREMENTS

We want to build, “above” an approximate icosahedral 3D crystal, a 6D icosahedral crystal of Bravais lattice L , a lattice commensurable with the primitive G -invariant

lattice $P = \sqrt{2}\mathbb{Z}^6$ ($G = m\bar{3}5$ denotes the icosahedral group), and describe its crystallography in terms of the symmetry operations of the 3D approximant crystal. Atoms will be assumed to be located on the 6D hypercubic host lattice $\sqrt{2}\mathbb{Z}^6$. In the present construction, they are supposed to be 0-dimensional, that is, they are points to which a color is assigned, and any point without color does not exist as an atom. They will be distributed by orbits (in strata) under the action of a 6D space group having the icosahedral group as point group. L is therefore a G -invariant sublattice of $\sqrt{2}\mathbb{Z}^6$. Let us recall some features about G -invariant lattices in $\mathbb{R}^6$.

Projection method from $\mathbb{R}^6$

We refer to Duneau and Katz (1985) for more details. Let $\{e_i/i = 1 \text{ to } 6\}$ the basis of $\sqrt{2}\mathbb{Z}^6$ such that $\{e_i/\sqrt{2}/i = 1 \text{ to } 6\}$ is the canonical orthonormal basis of $\mathbb{R}^6$. Physical space, denoted by E , is the image of the projector $P_{\parallel}$ expressed by the now classical matrix, in the basis of the e_i 's

$$P_{\parallel} = 1/2\sqrt{5} \begin{pmatrix} \sqrt{5} & 1 & 1 & 1 & 1 & 1 \\ 1 & \sqrt{5} & 1 & -1 & -1 & 1 \\ 1 & 1 & \sqrt{5} & 1 & -1 & -1 \\ 1 & -1 & 1 & \sqrt{5} & 1 & -1 \\ 1 & -1 & -1 & 1 & \sqrt{5} & 1 \\ 1 & 1 & -1 & -1 & 1 & \sqrt{5} \end{pmatrix} \quad (1)$$

The orthogonal projector is denoted by $P_{\perp} = Id - P_{\parallel}$. The inflation operator is $T = \tau P_{\parallel} - 1/\tau P_{\perp}$, satisfying $T^2 = T + 1$, and belongs to the collection of quasidilations $T_{\lambda\mu} = \lambda P_{\parallel} + \mu P_{\perp}$, λ, μ real numbers, which commute with the action of the icosahedral group G . We know that there are three classes of icosahedral Bravais lattices in $\mathbb{R}^6$ (Levitov and Rhyner, 1988): the primitive simple cubic: $P = \sqrt{2}\mathbb{Z}^6$, the face-centered cubic: $F = \{\sum n_i e_i \in P / \sum n_i \text{ even}, i = 1 \text{ to } 6\}$, the body-centered cubic: $I = P \cup (P + \Delta/2)$, where $\Delta = \sum e_i$, for $i = 1 \text{ to } 6$. The generating sets $\{(A5/a5), (A3/a3), (A2/a2)\}$ for each icosahedral space group are also characterized by these authors and we know that they are the only two (classes of) space groups for P , two for F , and only one for I , which is symmorphic. Here, we do not consider the icosahedral group 235 since, up to now, experimentally, we have only observed, by Convergent Beam Electron Diffraction, the non crystallographic point group $m\bar{3}5$ in the i -phase (Tanaka, Terauchi, Suzuki, Hiraga and Hirabashi, 1987). There are therefore a maximum of five 6D icosahedral crystals which can be built from the approximate icosahedral structure from which we start.

Rational cuts

By a rational cut in $\mathbb{R}^6$, we mean a couple (E', L') constituted of a 3D subspace E' and a rank 3 sublattice L' in the lattice $E' \cap P$. It is determined by 18 integers n_{ji} ($i = 1 \text{ to } 3, j = 1 \text{ to } 6$) which represent the three 6-tuples of coordinates, in the basis of the e_i 's, of a chosen set of generating vectors (w_1, w_2, w_3) of L' , and 2 relations. Since we

want to interpret the lattice $P_{\parallel}(L')$ as the Bravais lattice of the approximant phase, we obtain additional conditions for the n_j 's due to the metric relations imposed on their projections: $u_i = P_{\parallel}(w_i)$, by $P_{\parallel}$. For cubic structures, we have ten relations in addition. The first two relations arise from the linear independence conditions in E between the u_i 's. They can be automatically satisfied when the other relations between the n_{ji} 's are imposed by the metric tensor of the Bravais lattice of the 3D approximant crystal "c".

Let us denote by $P'_{\parallel}$ the rank 3 orthogonal projector of image E' . Expressed in the e_i 's, it has rational coefficients as well as the perpendicular projector $P'_{\perp} = yId - P'_{\parallel}$. Its orthogonal space in $\mathbb{R}^6$ is designated by $E'_{\perp}$ and is such that $E'_{\perp} \cap P$ is a rank 3 lattice. We call as usual $\{t_1, t_2, t_3\}$ a basis of $E'_{\perp} \cap P$. We note that $P'_{\parallel}(P)$ is discrete in E' and is a lattice containing L' . By irrational projection by $P_{\parallel}$, we obtain a lattice in physical space, namely $P_{\parallel}P'_{\parallel}(P)$, which will contain the Bravais lattice of the crystal, $P_{\parallel}(L')$. We call d the finite index $(P_{\parallel}P'_{\parallel}(P):P_{\parallel}(L'))$. We will make extensive use of both lattices in the construction method given below.

From the viewpoint of Fourier transforms, when we set a suitable band, around E' , to select nodes of P inside it and project the whole collection of these selected nodes on E by $P_{\parallel}$, we obtain a periodic approximant tiling for which the Fourier transform exhibits pseudoicosahedral symmetry if and only if E' is close to E (Verger-Gaugry, 1988). Moreover, in Fourier space, exact icosahedral positions, for each intense Bragg peak, can be defined collectively and distortions with respect to them can be indexed by 6-tuples of integers in the basis of a G-invariant lattice. Here, we do not want to work with tilings. However, because of the continuity of the factor structure of the approximant crystal with the atom positions in the unit cell, we can keep the same result in mind and state as a basic principle that 3D approximant crystals correspond to rational cuts (E', L') which are close to physical space in $\mathbb{R}^6$.

Proximity between E' and E is measured above the generating set $\{u_1, u_2, u_3\}$ by three rational numbers $p_i/q_i = 1/2 - 5N_{2i}/4N_{1i}$, for $i = 1$ to 3. Here, we have set $w_i = \sum n_{ji}e_j$, for $j = 1$ to 6, with $i = 1$ to 3; N_{2i} = the sum of the six squares n_{ji}^2 and

$$N_{1i} = n_{1i}(-n_{2i} - n_{3i} - n_{4i} - n_{5i} - n_{6i}) + n_{2i}(-n_{3i} + n_{4i} + n_{5i} - n_{6i}) \\ + n_{3i}(-n_{4i} + n_{5i} + n_{6i}) + n_{4i}(-n_{5i} + n_{6i}) - n_{5i}n_{6i}.$$

For each $i = 1$ to 3, we have the equivalence: $\|w_i - u_i\| \ll 1 \Leftrightarrow 0 < \tau - p_i/q_i \ll 1$. These quantities express the angular deviations between E' and E above the basis of the Bravais lattice, in $\mathbb{R}^6$. For cubic structures, they reduce to only one value, that we currently denote by p/q . They commonly take rational values with Lucas numbers, and not with Fibonacci numbers: $3/1, 7/4, 18/11, \dots$. The proximity to the golden mean is an indication of a "good" rational cut.

Exact compatibility of the approximant crystal with the projection method

For any x with rational coordinates in the e_i 's, let us denote by U the 6×1 column vector of its coordinates in the e_i 's. We have (Verger-Gaugry, 1990):

$$\|P_{\parallel}(x)\|^2 = {}^tU \cdot U + 1/\sqrt{5}({}^tU(2T - Id)U) \quad (2)$$

Square norms of such x_{ij} and therefore scalar products between such elements belong to the quadratic field $\mathbb{Q}(\sqrt{5})$. On the other hand, let us call C the metric tensor of the metric tensor of the Bravais lattice relative to the basis $\{v_1, v_2, v_3\}$ of the approximant 3D crystal living in E . We divide the metric tensor C by the square norm $\|v_1\|^2$ and set $C1 = C/\|v_1\|^2$. We have $\|y\|^2 = \|v_1\|^2(YC1'Y)$, where Y denotes the column vector of the coordinates of the element y in the v_i 's. $\|v_1\|$ is a lattice parameter, and is given experimentally by a rational number times a unit length, traditionally nm, allowing to consider E , 3D Euclidean space, as our physical space endowed with the SI units system of reference. A consequence is that $C1$ is normalized, that is, without units. We now make the correspondence with the projection method. We set:

$$K = \mathbb{Q}(\|v_i\| \|v_j\| \cos(v_i, v_j) / \|v_1\|^2) \quad 1 \leq i, j \leq 3 \quad (3)$$

and say that the approximant crystal is exactly compatible with the projection method from $\mathbb{R}^6$ when the field $K = \mathbb{Q}$ or $\mathbb{Q}(\sqrt{5})$. K is stable under basis changes since C is changed into $A'CA$, where A is 3×3 integral unimodular. K is a constant of the lattice referred to $\{v_1, v_2, v_3\}$. If we take $\|v_2\|$ or $\|v_3\|$ instead of $\|v_1\|$, we obtain the same field K with another equivalent metric transfer between the 3D Euclidean space and physical space.

For cubic structures, $K = \mathbb{Q}$. For hexagonal structures for which c/a belongs to $\mathbb{Q}(\sqrt{5})$, we have $K = \mathbb{Q}(\sqrt{5})$. For the rhombohedral structure used to describe $i\text{-Al}_6\text{CuLi}_3$, of space group $R\bar{3}m$, whose unit cell is a prolate rhombohedron inflated by τ^3 , we have cosines all equal to $1/\sqrt{5}$, and $K = \mathbb{Q}(\sqrt{5})$ (Audier and Guyot, 1988). It is clear that, for an arbitrary phase, an approximate compatibility of its Bravais lattice with the projection method from $\mathbb{R}^6$ can easily be reached when K is bigger than $\mathbb{Q}(\sqrt{5})$, since lattice parameters and their angular relations are defined only within a certain precision, and that $\mathbb{Q}(\sqrt{5})$ is dense in $\mathbb{R}$. We can therefore assume in the following that the approximant crystal has a Bravais lattice exactly compatible with the projection method. If this is the case, the modelization of the lattice, as L , is unique up to a normalization factor in $\mathbb{Q}$, therefore in $\mathbb{Z}$.

CONSTRUCTION OF THE 6D ICOSAHEDRAL CRYSTAL

Icosahedral completion of the atomic decoration

We consider (Tamura, 1989; Verger-Gaugry, 1990) that an approximant crystalline "cousin" phase "c" of an icosahedral quasicrystal "i", such as $R - \text{Al}_5\text{CuLi}_3$, $\alpha - \text{AlMnSi}$, $c - \text{Ti}_2\text{Ni}$, is defined by: 1) in reciprocal space, a distribution of intense Bragg peaks which almost follows icosahedral symmetry, without respecting it since diffraction spots are located on a lattice, 2) in real space, the existence of an important density of CN12 sites, empty or occupied, as in many Frank-Kasper phases, about which pseudo-icosahedral units of more or less important diameters lie. In the structure, these units are strongly imbricated one into the others.

Let us call "6-c" the 6D icosahedral crystal assumed to be above "c" and "i" in the same system. We build a part of it as follows: if $x \in P$ is such that $P_{\parallel} P'_{\parallel}(x)$ is the position of an atom, the point x takes the color of this atom and "becomes" an atom. The quantity d , index of the Bravais lattice $P'_{\parallel}(L')$ of "c" in the lattice $P_{\parallel} P'_{\parallel}(P)$, represents the maximal number of host atomic sites for the atoms in the unit cell. Only a subset of $P_{\parallel} P'_{\parallel}(P)$ is effectively constituted by the (real) atom positions. Many nodes are empty. This coloration of the primitive lattice P in $\mathbb{R}^6$ depends upon the choice of a "good" rational cut (E', L') . We will indicate below how to choose it, we think, in the best way, according to local order in "c". After this first step, we thus obtain a 6D crystal above "c". However, it has no reason to be invariant under the action of an icosahedral space group. On the other hand, we need to have a 6D icosahedral crystal "6-c" above "i" since, by irrational projection $P_{\parallel}$ on E , we should obtain, with a suitable band algorithm, an aperiodic arrangement of atoms, called "i"-phase, which should exhibit in the Fourier transforms G -invariance symmetry and inflation rules deduced from the symmetry operations of "6-c". To force this compatibility, we are then bound to view the 6D icosahedral crystal "6-c" as the completion, under an icosahedral space group F_6 , of the 6D crystal that we have just built (by lifting up the atomic decoration of the approximant crystal by $P_{\parallel} P'_{\parallel}$). F_6 is not arbitrary; it is such that its translation vectors lattice T_6 is compatible with the existing atomic decoration as we now explain it. If DL (for Decoration Lattice) represents the lattice generated by $\{t_1, t_2, t_3\}$ and L' , of covolume m (=the index $(P:DL)$) with respect to P , then mP (the lattice P multiplied by m), which is G -invariant, is a sublattice of DL (Cohn, 1980). DL bears in its unit cell all the information about the atomic decoration of the approximant structure by construction and mP is compatible with this decoration. We now take GDL the densest and greatest G -invariant lattice included in DL , containing mP . We take $T_6 = GDL$. It is the closest G -invariant lattice compatible with the atomic decoration. T_6 is equivalent to one of the three lattices P, F, I . There are therefore one possibility for F_6 , if T_6 is equivalent to I , and two in the other cases. The full icosahedral crystal "6-c" is thus obtained by completion. There is no accumulation of atoms in bounded regions of $\mathbb{R}^6$ since all atoms live in the (discrete) lattice P .

We then say that the rational projector $P'_{\parallel}$ is compatible with the 6D icosahedral crystal "6-c", of space group F_6 , when, after completion by the suitable space group F_6 , all nodes of P are affected by at most one color. In other words, this construction makes sense when there is no superposition of atoms.

Splitting of atom orbits and Wyckoff positions

Let us write the exact sequences characterizing the respective space groups F_3 and F_6 of the crystals "c" and "6-c". The subscripts indicate only the dimension of the Euclidean space in which the crystals live. We have:

$$1 \rightarrow T_3 \rightarrow F_3 \rightarrow R_3 \rightarrow 1 \quad (4)$$

$$1 \rightarrow T_6 \rightarrow F_6 \rightarrow R_6 \rightarrow 1 \quad (5)$$

with $R_6 = G$. They are not necessarily splitted, except if $T_6 = I$ for the second one. Relations between F_3 and F_6 are not obvious. In the previous section, we have lifted up the elements of $T_3 = P_{||}(L')$ on L and then on GDL. Now, if we try to lift up the symmetry operations of R_3 directly on P to obtain orthogonal elements in R_6 , we will not succeed. Indeed, the elements of R_3 leave invariant the lattice T_3 , and by the lifting up process, should arise from space group elements, with zero translation part, preserving L' and therefore E' , by linearity. However, the only subspaces stable under the action of G in $\mathbf{R}^6$ are E and its orthogonal $E_{\perp}$. We therefore cannot hope to embed the point group R_3 , or some of its subgroups, in the icosahedral group G .

If we want to envisage differently the icosahedral atomic completion, we consider, for each atom orbit j in "c" modulo its Bravais lattice R_3 , the set S_j of all translation vectors having their extremity and their origin in j . Only a finite number of values can be taken by j . By construction, S_j is included in $P_{||}P'_{||}(P)$. All the translations are then lifted up in P , not uniquely. All the translations vectors thus generated in $\mathbf{R}^6$ are not all equivalent under the action of F_6 , and their classes, in finite number, give the 6D atom orbits in "6-c". Above an atom orbit in a given Wyckoff position, we therefore obtain several atoms orbits in several Wyckoff positions, giving rise to a discrete covering of the structure. The union of all these 6D atom orbits under F_6 gives the "6-c" icosahedral crystal.

Optimal embedding of the approximant crystal in $\mathbf{R}^6$

Optimality is defined with respect to local environments about CN12 sites in the 3D approximant "cousin" crystal. To embed, in an optimal way, an approximant crystal in physical space E amounts to fitting, by the least squares method, a perfect icosahedron (or a perfect icosahedral motif) to the whole set of pseudo-icosahedral clusters of the "c" phase, moved by translation in such a way that all their centers coincide in one point, the origin for instance (Figure 1).

Figure 1 Fit of a perfect icosahedron with respect to a collection of distorted icosahedra: a) all pseudo-icosahedral clusters are moved by translation; the origin becomes their common center. b) the position of the perfect icosahedron provides the directions of the $e_{i||}$'s, and therefore the action of the icosahedral group in physical space referred to the basis of the Bravais lattice of the approximant structure.

We have applied this method to the following approximant structures: α -AlMnSi, R -Al₅CuLi₃, β -AlMnSi and c -Ti₂Ni (Tamura, Verger-Gaugry and Barbier, 1990). It decomposes into three steps: 1) analysis of local order in the approximant structure, 2) fit of a perfect icosahedron to the whole set of distorted icosahedra, or to certain families of distorted icosahedra, 3) calculation of a rational cut. The first two phases were extensively studied (Gratias, 1988); the third one was reconsidered by Le Lann (1990) as an approximant structure; the last one was studied by Zhang *et al.* (1986) and François *et al.* (1990).

1) In these approximant phases, pseudo-icosahedral clusters are strongly interpenetrated one into the others, but define, globally, by averaging, one, two or four icosahedral orientations (each of them with a very small standard deviation of the distribution); these clusters are therefore classified by families:

α -AlMnSi: (space group $Pm\bar{3}$, $a = 1.268$ nm, 138 atoms per unit cell); the reference cluster is the double Mackay icosahedron. Pseudo-icosahedral character propagates far beyond third neighbours to give an hexecontahedron, an icosahedron and finally an icosidodecahedron. In this phase, pseudo-icosahedral clusters gather into only *one* orientational family.

R -Al₅CuLi₃: (space group $Im\bar{3}$, $a = 1.389$ nm, 160 atoms per unit cell); this structure was redetermined by Audier *et al.* (1988), to improve the previous results of Cherkashin, Kripyakevich and Oleksiv (1964). As in α -AlMnSi, pseudo-icosahedral clusters are almost perfect. Pseudo-icosahedral order propagates up to 6th neighbours with quasiregular polyhedra. Pseudo-icosahedral units constitute only *one* orientational family.

β -AlMnSi: (space group $P6_3/mmc$, $a = 0.275$ nm, $c = 0.77$ nm, 26 atoms per unit cell); distorted icosahedra are centered on the Si atoms located at (0, 0, 0) and (0, 0, 1/2). There are *two* orientational families in this structure.

c -Ti₂Ni: (space group $Fd\bar{3}m$, $a = 1.1278$ nm, 96 atoms per unit cell); 50% of atomic sites are surrounded by pseudo-icosahedral clusters. This phase contains *four* (physically relevant) orientational families of such clusters, around Ni(e) atoms. The succession of pseudo-icosahedral shells having Ni (e) centers and Ti (c) centers is given in Figures 2a and 2b.

2) Optimization: all distorted icosahedra in the unit cell of the structure, or only those belonging to the same family are now centered at the origin. By dilation, all the vertices of the icosahedra are placed on a sphere S centered at (0, 0, 0). We take its radius equal to the average value of the distances of the common center to the vertices of the various distorted icosahedra. If (ξ_j, ψ_j, ζ_j) denotes the current position of the j th vertex of the perfect rigid icosahedron inscribed in S , if (x_{ij}, y_{ij}, z_{ij}) is the coordinates of the j th vertex of the i th distorted icosahedron viewed on S by dilation from the origin, we minimize the following function by the least squares method (we have used the Brent method to invert the Jacobian matrix):

$$DEV = \sum_i \sum_j ((x_{ij} - \xi_j)^2 + (y_{ij} - \psi_j)^2 + (z_{ij} - \zeta_j)^2) \quad (6)$$

We just indicate the results for c -Ti₂Ni and refer to Tamura *et al.* (1990) for further

Figure 2 a) Decomposition of the pseudo-icosahedral cluster centered on a Ni(e) atom in $c\text{-Ti}_2\text{Ni}$, up to third neighbours. b) Sequence of pseudo-icosahedral shells, up to 6th neighbours, in a pseudo-icosahedral cluster centered on a Ti(c) atom in $c\text{-Ti}_2\text{Ni}$.

details about the other phases. Applied to all distorted icosahedra, we obtain very large angular deviations, up to 62 degrees for one vertex, between the optimal position of the icosahedron and the distribution (Table 1a). Applied to each family, it provides a much better fit (Table 1b and 2). Angular deviations are much smaller, less than 3 degrees per vertex. We observe that the position of the perfect icosahedron in the family of distorted icosahedra used in Table 2 gives exactly the orientation relationships put into evidence by TEM by Zhang *et al.* (1985) (Figure 3). The present approach predicts other orientation relationships between *i*-Ti₂Ni and *c*-Ti₂Ni with an optimized action of the icosahedral group in physical space defined by the position of this icosahedron.

3) Calculation of a rational cut: the above fit provides the orientation of the e_{ij} 's in the basis of the Bravais lattice of the approximant structure. These vectors are determined, up to a normalization factor k , that we take in $\mathbf{Z}[\tau] = \{a + b\tau/a, b \in \mathbf{Z}\}$. The 18 integers n_{ij} , coordinates of the vectors w_i of the basis of L' , characterizing the rational cut (E', L') are then calculated by optimization under the following conditions:

i) the u_i 's constitute the basis of the Bravais lattice of the approximant structure, with correct angular relations and lattice parameters ratios.

Table 1 a) Coordinates of the 12 vertices of the perfect icosahedron after optimization, in the basis of the Bravais lattice. The distribution of distorted icosahedra is constituted of 32 icosahedra around Ni(e) atoms, and 16 icosahedra around Ti(c) atoms in the approximant crystal *c*-Ti₂Ni. To obtain the precision on these coordinates, the cumulative derivatives have to be multiplied by the precision, currently 10^{-3} , on the position parameters. b) The distribution of distorted icosahedra is restricted to one orientational family in *c*-Ti₂Ni. After optimization, the angular deviations per vertex do not exceed 3 degrees. The precision on these coordinates is obtained by multiplying the cumulative derivatives by 10^{-3} (Ni(e) centers).

vertex	X	Y	Z	angular deviation (deg.)
1	-0.59133E - 01	-0.25922E - 01	-0.96401E + 00	10.76
7	0.59133E - 01	0.25922E - 01	0.96401E + 00	9.90
2	0.34382E + 00	0.66435E + 00	-0.66365E + 00	4.23
8	-0.34382E + 00	-0.66435E + 00	0.66365E + 00	9.08
3	-0.68473E + 00	0.47774E + 00	-0.55037E + 00	21.63
9	0.68473E + 00	-0.47774E + 00	0.55037E + 00	24.23
4	-0.80355E + 00	-0.52930E + 00	-0.27229E + 00	24.23
10	0.80355E + 00	0.52930E + 00	0.27229E + 00	15.01
5	0.15156E + 00	-0.96507E + 00	-0.21370E + 00	14.30
11	-0.15156E + 00	0.96507E + 00	0.21370E + 00	11.36
6	0.86068E + 00	-0.22736E + 00	-0.45557E + 00	16.54
12	-0.86068E + 00	0.22736E + 00	0.45557E + 00	62.44
average angular deviation = 0.23047E + 02				
standard deviation of the distribution ($0 \leq e \leq 1$): 0.39954E + 00				
cumulative derivatives of outputs versus input data				
1-7	0.45199E + 00	0.47936E + 00	0.13077E + 00	
2-8	0.43840E + 00	0.38922E + 00	0.36166E + 00	
3-9	0.33906E + 00	0.43747E + 00	0.42811E + 00	
4-10	0.27611E + 00	0.38274E + 00	0.46186E + 00	
5-11	0.43296E + 00	0.13822E + 00	0.49590E + 00	
6-12	0.23749E + 00	0.50793E + 00	0.41648E + 00	

Table 1 b)

vertex	X	Y	Z	angular deviation (deg.)
1	0.36870E - 01	-0.70663E + 00	-0.70663E + 00	1.25
7	-0.36870E - 01	0.70663E + 00	0.70663E + 00	1.25
2	0.29269E + 00	0.29269E + 00	-0.91031E + 00	2.97
8	-0.29269E + 00	-0.29269E + 00	0.91031E + 00	2.97
3	-0.70663E + 00	0.36870E - 01	-0.70663E + 00	1.25
9	0.70663E + 00	-0.36870E - 01	0.70663E + 00	1.25
4	-0.70663E + 00	-0.70663E + 00	0.36870E - 01	1.25
10	0.70663E + 00	0.70663E + 00	-0.36870E - 01	1.25
5	0.29269E + 00	-0.91031E + 00	0.29269E + 00	2.97
11	-0.29269E + 00	0.91031E + 00	-0.29269E + 00	2.97
6	0.91031E + 00	-0.29269E + 00	-0.29269E + 00	2.97
12	-0.91031E + 00	0.29269E + 00	0.29269E + 00	2.97
average angular deviation = 0.22769E + 01				
standard deviation of the distribution ($0 \leq e \leq 1$): 0.39737E - 01				
cumulative derivatives of outputs versus input data				
1-7	0.49695E + 00	0.28734E + 00	0.28734E + 00	
2-8	0.43334E + 00	0.43334E + 00	0.20584E + 00	
3-9	0.28734E + 00	0.49695E + 00	0.28734E + 00	
4-10	0.28734E + 00	0.28734E + 00	0.49695E + 00	
5-11	0.43334E + 00	0.20584E + 00	0.43334E + 00	
6-12	0.20584E + 00	0.43334E + 00	0.43334E + 00	

Table 2 Another orientational family of distorted icosahedra, around Ni(e) atoms, in c -Ti₂Ni, is used. The resulting angular deviations are small, less than 3 degrees. The coordinates of the vertices of the perfect icosahedron, together with their calculated precision equal to the cumulative derivatives times 10^{-3} , are fully compatible with the experimental results given in Figure 3.

vertex	X	Y	Z	angular deviation (deg.)
1	0.70663E + 00	0.70663E + 00	0.36870E + 00	1.25
7	-0.70663E + 00	-0.70663E + 00	-0.36870E - 01	1.25
2	0.91031E + 00	-0.29269E + 00	0.29269E + 00	2.97
8	-0.91031E + 00	0.29269E + 00	-0.29269E + 00	2.97
3	0.29269E + 00	0.29269E + 00	0.91031E + 00	2.97
9	-0.29269E + 00	-0.29269E + 00	-0.91031E + 00	2.97
4	-0.29269E + 00	0.91031E + 00	0.29269E + 00	2.97
10	0.29269E + 00	-0.91031E + 00	-0.29269E + 00	2.97
5	-0.36870E - 01	0.70663E + 00	-0.70663E + 00	1.25
11	0.36870E - 01	-0.70663E + 00	0.70663E + 00	1.25
6	0.70663E + 00	-0.36870E + 01	-0.70663E + 00	1.25
12	-0.70663E + 00	0.36870E - 01	0.70663E + 00	1.25
average angular deviation = 0.22770E + 01				
standard deviation of the distribution ($0 \leq e \leq 1$): 0.39738E - 01				
cumulative derivatives of outputs versus input data				
1-7	0.28734E + 00	0.28734E + 00	0.49695E + 00	
2-8	0.20584E + 00	0.43334E + 00	0.43334E + 00	
3-9	0.43334E + 00	0.43334E + 00	0.20584E + 00	
4-10	0.43334E + 00	0.20584E + 00	0.43334E + 00	
5-11	0.49695E + 00	0.28734E + 00	0.28734E + 00	
6-12	0.28734E + 00	0.49695E + 00	0.28734E + 00

Figure 3 Superimposition of the stereograms of the *i* phase (in black) and the crystalline phase *c*-Ti₂Ni (in white), with $\bar{5}/c[110]$ and $\bar{2}/c[1\bar{1}0]$, (after Zhang and Kuo, 1986).

ii) the n_{μ} 's must remain small, in order to obtain as small as possible d values. Indeed, the density d of host atomic sites should remain a physically interesting quantity and a bound for it, that we can take equal to 10^9 , is directly given by the precision, currently 10^{-3} , on the parameters positions. The lattice $P_{\parallel}P'_{\parallel}(L)$ should cover all the atomic sites in E , by coincidence (Verger-Gaugry, 1990), with a minimal density d .

iii) E' must be as close as possible to E , that is we minimize the p_i/q_i values, in order to obtain them as close as possible to the golden mean.

PHASE TRANSITIONS BETWEEN THE I-PHASE AND THE APPROXIMANT CRYSTALLINE PHASE

In order to obtain the icosahedral phase *i* in physical space, it is now commonly admitted that we should make an irrational selection by a band algorithm (Gratias, 1988) applied to the 6D icosahedral object and project it irrationally on E ; here, in the present framework, it would be "*i*" from "*6-c*". The geometry of the band is made for obtaining no interpenetration of the atoms after projection on E and keeping a local maximal compacity. We would like to show now that the scheme is similar

to obtain the approximant 3D crystal from the "6-c" phase, but, here, there is no more band algorithm.

Indeed, before the completion of the atomic decoration under a suitable space group, the 6D crystal looks like a union of colored parallel affine 3D lattices, all included in the primitive lattice P . This stratified structure arises directly from the rational cut used, since their common orientation is $E'_\perp$, the orthogonal space to E' . The completion admits therefore a converse operation, given a 6D icosahedral crystal "6-c": in order to obtain the approximant 3D crystal by a rational cut (E' , L) and the double projection $P_\parallel P'_\parallel$, we "read diagonally" inside the 6D icosahedral crystal "6-c" so as to select all the parallel colored 3D affine lattices in P of common orientation $E'_\perp$. All other atoms are not selected. We then project these 3D parallel lattices on E' and finally on E (by $P_\parallel P'_\parallel$). Each of them provide only *one* atom in E by construction and we recover the 3D approximant crystal.

We therefore view such irrational and rational "selection + projection" processes as two similar ways to visualize, partially in both cases, the same 6D icosahedral crystal. We obtain in this way two visibility modes of the same 6D object.

The rational selection mode described above is directly linked to arithmetic selection rules as far as we use rational cuts with rational projectors. We can restrict these rules to each atom orbit (each occupied Wyckoff position), to obtain the partial occupation of the concerned color, and correlate this occupation to the occupation of the corresponding atom orbit in the 3D approximant phase. This provides a generalization of the chemical order parameters (Portier and Gratias, 1981) which are currently defined in the order-disorder phase transitions (Verger-Gaugry *et al.*, 1990).

CONCLUSION

The phase transition between the i phase and its "cousin" phase can be viewed in two ways: either by coherent collective displacements in 3D real Euclidean space with atoms diffusion only on small distances bounded by a constant (notion of bounded one-to-one moves of Duneau and Oguey, (1990)), or by the creation of a suitable rational cut in $\mathbf{R}^6$, compatible with the icosahedral 6D crystal, such that the 6D crystal, which was projected irrationally, is now projected rationally. We assume that both are strictly equivalent; this gives a means to algebraize the phases transition between both phases by optimal choices of rational cuts in $\mathbf{R}^6$ and a change of "selection + projection" mode.

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